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Neutron Multiplicity Assay of Impure Materials Using Four Different Neutron Counters

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Abstract

During an advanced IAEA inspector training course given at Los Alamos in November, 1997, the opportunity existed for an intercomparison study of various neutron detectors to quantify measurement performance using pure and impure plutonium oxide and mixed uranium-plutonium oxide (MOX) items. Because of the cost of counters designed specifically for multiplicity analysis, it was desired to explore the limits of other, less costly and less efficient detectors. This paper presents and intercompares neutron coincidence and multiplicity assay performance for five detectors, which vary widely in detection efficiency. Eight pure plutonium oxide and twelve impure plutonium oxide and MOX working standards were used in the study.

INTRODUCTION

During the past twenty years, a new nondestructive assay (NDA) method for plutonium, called passive neutron multiplicity counting (PNMC), has been developed as an extension of passive neutron coincidence counting (PNCC). PNMC was developed for assay of impure, plutonium bearing materials. The new technique has led to the design and fabrication of a new generation of instruments called neutron multiplicity counters (NMCs). The development of new neutron counters has been accompanied by advances in data processing electronics, analysis algorithms, and data collection software. Development activities have been supported primarily by the DOE Office of Safeguards and Security, Technology Development Branch. In recent years, implementation of the new method has supported application of IAEA Safeguards at two US facilities, Hanford and Rocky Flats, for materials excess to national security needs. The DOE Office of Nonproliferation and National Security and the Program of Technical Assistance to IAEA Safeguards have supported implementation of PNMC for verification of excess US weapons materials.

In November, 1997, a special training course in neutron multiplicity counting and assay was held at Los Alamos for twelve experienced IAEA inspectors. The inspectors were teamed in five groups, each using a different neutron coincidence or multiplicity counter. The scope of the course included characterization and calibration of the five counters, and assay of both pure and impure plutonium oxide items. The five neutron counters used were the High-Level Neutron Coincidence Counter (HLNC),[1] the Active-Well Coincidence Counter (AWCC) [2,3] - two units, operated in passive mode, the 3-Ring Multiplicity Counter (3RMC),[4,5,6] and the the 5-Ring Multiplicity Counter (5RMC).[7] These counters vary widely in detection efficiency, from 17.1% for the HLNC to 54% for the 5RMC.

The data set produced during the training exercise produced interesting results. The assay accuracy and precision of the various counters can be directly compared for multiplicity analysis, conventional coincidence analysis, and multiplication-corrected coincidence analysis. This paper provides an intercomparison of the results obtained, along with conclusions.

ANALYSIS METHODS

Neutron coincidence counting (NCC) is used extensively for NDA of bulk nuclear material. Detector heads use ^{3}He proportional counters moderated by polyethylene. Fast neutrons from the sample are slowed in the polyethylene and captured in the ^{3}He. NCC is useful primarily because fast neutrons penetrate the sample well and because time-correlated neutrons are directly related to the fission process and thus to the nuclear material content. Special pulse-processing electronics enable separation of time-correlated from random neutron pulses. NCC is used in passive mode (PNCC) to assay the even, spontaneously fissioning isotopes of plutonium (^{238}Pu, ^{240}Pu, and ^{242}Pu) and in active mode to assay induced-fissionable isotopes, primarily ^{235}U, in bulk samples. In passive mode, no external neutron sources are required. In active mode, AmLi sources are typically used to induce fissions in ^{235}U.

Passive NCC is well suited for measuring dense plutonium-bearing materials, such as pure metal and oxide, over a wide range of masses. Precision and accuracy are much better for pure materials than for scrap. For pure plutonium oxide, there are two primary sources of neutrons and one secondary source. The two primary sources are spontaneous fission and (a,n) reactions. Several plutonium isotopes decay by prolific emission of alpha particles, some of which undergo reactions with oxygen to produce single neutrons. The secondary source of neutrons in plutonium oxide is fast-neutron induced fission, occurring in all plutonium isotopes. For pure plutonium metal, the (a,n) neutron source is zero because of the absence of low-Z elements.

For plutonium scrap, all three sources of neutrons exist as with plutonium oxide; however, the (a,n) component is calculable for pure oxide, but not for scrap, unless an impurity analysis is available. Table I below summarizes primary and secondary neutron sources and their origins for plutonium metal, oxide, and scrap.

Table I. Primary and Secondary Neutron Sources from Plutonium Metal, Oxide, and Scrap

	Spontaneous Fission	Induced Fission	(a,n) Reactions
Pure Metal	^{238}Pu, ^{240}Pu, and ^{242}Pu	^{238}Pu thru ^{242}Pu	none
Pure Oxide	^{238}Pu, ^{240}Pu, and ^{242}Pu	^{238}Pu thru ^{242}Pu	oxygen
Impure Oxide and Scrap	^{238}Pu, ^{240}Pu, and ^{242}Pu	^{238}Pu thru ^{242}Pu	oxygen plus other low-Z impurities, e.g., fluorine

For PNCC assay of plutonium materials, calibration is performed using physical standards to determine a curve with the real coincidence count rate *R* plotted versus the effective ^{240}Pu mass. The effective ^{240}Pu mass contains contributions from ^{238}Pu and ^{242}Pu: it is the mass of ^{240}Pu that would give the same neutron coincidence response as that obtained from all the even plutonium isotopes in the sample.

If there were no induced fission in the sample, the calibration curve of *R* versus the effective ^{240}Pu mass would be linear and the effects of variations in impurities, geometry, and density would be relatively unimportant. However, the existence of induced fission (multiplication) in the sample causes these effects to be important considerations in preparing standards for and calibrating TNCCs.

Calibration Curve - Figure 1 shows two calibration curves for a set of pure plutonium oxide standards. The upper *R* curve is uncorrected. The lower curve is corrected for multiplication using a method described below. The lower curve contains contributions from spontaneous fission only.

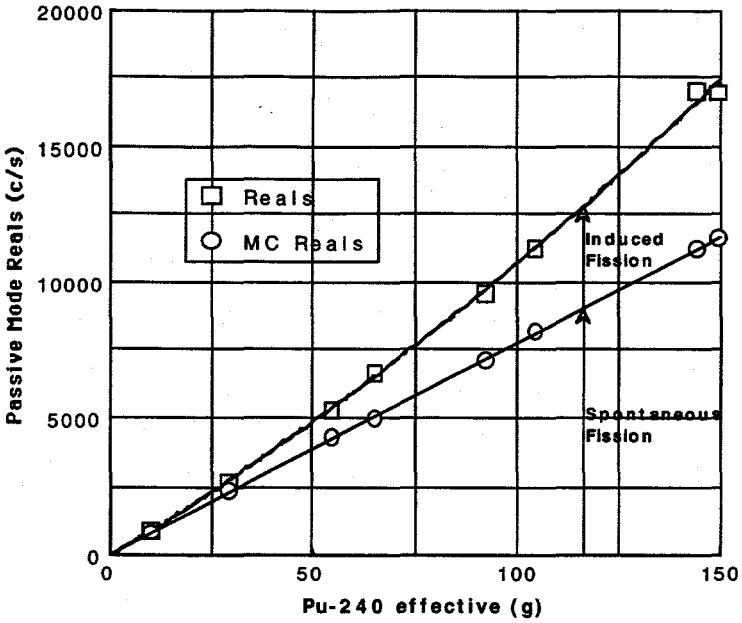

Fig. 1. Calibration curves for PNCC of pure plutonium oxide. Upper curve is the real coincidence count rate (Reals) versus the effective ^{240}Pu mass. Lower curve (MC Reals) is corrected for induced fission or multiplication.

Fissions may be induced in the item to be assayed by primary neutrons born in spontaneous fission and (a,n) reactions or by secondary neutrons from other induced fissions. For a given effective ^{240}Pu mass, the number of induced fissions, and therefore the contribution to *R*, depends on (a,n) reactions in the sample and also on plutonium isotopic composition, geometry, and density. The self-

multiplication of the item (ratio of primary plus secondary to primary neutrons) increases with plutonium mass and density.

"Known-a" - For pure plutonium oxide, or impure plutonium oxide with known impurities, there are three sources of neutrons, but the ratio of (a,n) to spontaneous fission neutron production (a) is known from plutonium isotopics ratios and nuclear data. Therefore, in this case, there are two unknown neutron source rates and two measured parameters, *R* and *T*. Solving these two equations results in a "multiplication-corrected" *R* that is used for calibration and assay. This "known-a" method is preferable to the uncorrected-*R* calibration for pure materials or for impure materials with known impurities because variations in *R* due to variations in induced fission are removed. The upper curve of Fig. 1. contains these variations. Precision is improved with the "known- a" method because both *R* and *T* are used in the analysis, and the measurement of *T* is more precise than *R*. For most impure plutonium materials for assay, however, impurity levels are unknown and the method is usually restricted to pure materials (metal and oxide). In cases where geometry and density are constant for a set of impure items, a calibration of multiplication *M* versus effective ^{239}Pu mass can be used to eliminate one of the three unknown neutron source rates. This "known-*M*" method is useful in some cases, but is not used in this paper.

Multiplicity Analysis - PNMC detectors are similar in design and construction to PNCC detectors: they both use polyethylene-moderated ^{3}He proportional counters. However, multiplicity counters are designed to maximize counting efficiency and minimize neutron energy dependence and deadtime. In addition to measuring the total count rate *T* ("singles") and the real coincidence count rate *R* ("doubles"), multiplicity counters also measure the third moment ("triples") of the neutron multiplicity distribution. While *T* and *R* are proportional to efficiency and the efficiency squared, respectively, the triples rate is proportional to the cube of the neutron counting efficiency. The desire to obtain the best precision of the triples measurement leads to high-efficiency detectors.

PNMC detectors were developed to provide three measured parameters for assay of impure plutonium-bearing materials. As mentioned previously, these materials have three sources of neutrons to be resolved: spontaneous fission, induced fission and (a,n) reactions. Multiplicity analysis involves solution of three equations for the singles, doubles, and triples in the three unknowns to yield the effective ^{240}Pu mass. The effects of unknown multiplication and (a,n) impurities are removed from the assay.

Physical standards for PNMC assay are not required in the usual sense. They are required to verify proper setting of a few detector parameters used in the multiplicity analysis. The concept of generating a calibration curve of instrument response to effective ^{240}Pu mass does not apply to PNMC assay.

Initial calibration of a PNMC detector can be done with a ^{252}Cf source alone. However, in practice, it is best to use a well-characterized pure or impure plutonium standard to verify detector parameters. The calibration can then be used to assay items with a wide range of impurity concentrations, plutonium isotopes, bulk density, fissile mass, and geometry.

CALIBRATIONS

A set of eight plutonium oxide standards ("LAO series"), with ~16 % ^{240}Pu mass fraction, ranging in mass from 60 to 870 g of plutonium, was prepared in 1983 for PNCC calibrations. The LAO series is well-characterized and considered "pure" in terms of low-Z contaminants and water (< 0.5%). The LAO set is well-characterized with regard to plutonium mass, isotopics, low-Z impurities, and moisture. The a value for the LAO series was ~0.47. The standards are doubly contained in welded steel food-pack cans.

^{252}Cf sources and the LAO series of standards were used for characterization and calibration of the five detectors including calibration curve, known-a, and multiplicity analysis methods. Table II below lists selected parameters for the five detectors as determined by the participants in the inspector training course.

Table II. Detector Parameters

Detector	Efficiency	Pre-delay (ms)	Gate Width (ms)	Doubles Gate Fraction	Triples Gate Fraction	Die-Away Time (ms)	Deadtime Coefficient A (1E-6)	Deadtime Coefficient B (1E-12)	Multiplicity Deadtime (ns)
HLNC	0.171	3	64	0.701	0.492	42.1	0.768	0.248	215
AWCC1	0.328	3	64	0.656	0.431	51.2	0.826	0.267	237
AWCC2	0.336	3	64	0.660	0.458	50.9	0.700	0.230	210
3RMC	0.425	3	64	0.631	0.414	55.0	0.315	0.102	90
5RMC	0.540	3	64	0.628	0.409	55.2	0.184	0.059	61

ASSAY RESULTS FOR PURE AND IMPURE PLUTONIUM OXIDE ITEMS

In addition to the 8 LAO items, 12 impure oxide items were available for assay. Two of the items are mixed uranium-plutonium oxide (MOX). The 12 items ranged in mass from 20 to 875 g Pu, and their a values ranged from 0.72 to 1.1. One impure item, with a mass of 60 g Pu, had an a value of 4.3. Each of the five inspector groups worked with a single detector, and performed assays of the pure (LAO series) and impure oxide items. Each group used three analysis methods for assay, i.e., calibration curve, known-a, and multiplicity. Count times were chosen to give the best statistical precision, while also trying to maximize the number of items assayed in the time allowed. The HLNC required the longest count times because of the lowest efficiency. The 5RMC required the shortest count times because of the highest efficiency. Assay results are summarized in Table III below. The table entries are %(D-A)/D where D is the declared value based on chemical analysis and weighing, and A is the assay value.

Table III. Summary Assay Results for Five Detectors and Three Analysis Methods

IAEA Inspector Group	Detector	Pu Oxide (# items)	Nominal Count Time (s)	Calibration Curve			Known Alpha			Multiplicity		
				Mean Bias (%)	Standard Deviation (%)	Precision (%)	Mean Bias (%)	Standard Deviation (%)	Precision (%)	Mean Bias (%)	Standard Deviation (%)	Precision (%)
1	5RMC	pure (8)	600	-0.15	1.67	1.69	-0.68	0.63	0.36	0.93	1.55	1.06
2	AWCC1	pure (4)	600	-1.40	1.52	1.26	-0.79	0.63	0.41	0.32	3.31	1.93
3	3RMC	pure (3)	300	-0.60	1.87	0.43	-0.17	0.75	0.26	-3.67	4.06	2.03
4	AWCC2	pure (4)	1000	-1.18	1.04	0.54	-1.33	0.60	0.26	-1.13	1.20	1.27
5	HLNC	pure (4)	1000	-0.70	1.30	0.50	-1.55	0.85	0.20	-1.02	1.66	3.18
1	5RMC	impure (11)	600	-7.78	9.19	2.15	-12.45	11.16	0.56	0.84	2.56	1.64
2	AWCC1	impure (8)	900	-9.48	10.95	1.72	-13.24	16.78	0.52	-0.39	1.96	1.76
3	3RMC	impure (9)	1200	2.26	17.95	1.47	-7.60	4.72	0.47	-2.08	2.45	1.74
4	AWCC2	impure (8)	1200	-9.27	11.09	0.85	-15.94	13.58	0.55	0.10	2.55	2.44
5	HLNC	impure (10)	1300	-8.35	11.30	0.68	-17.01	13.46	0.56	-1.14	3.50	4.48

For the LAO series of pure oxides, assay biases range from -1.40% to -0.15% for the calibration curve method, and from -1.55% to -0.17% for the known-a method. As expected, the precision of the known-a method is better than for the calibration curve method. The multiplicity method produced biases ranging from -3.67% to +0.93% for the pure oxides. The precision of the multiplicity assays is worse than for the calibration curve method, except for the 5RMC. This is due to the high efficiency of the 5RMC and hence its capability to more precisely measure triples than the other counters.

For the impure oxides, the calibration curve method produced biases from -9.48% to +2.26% - much worse than for the pure items. This is due to differences in (a,n) reactions in the sample and also differences in plutonium isotopic composition, geometry, and density, found when comparing the pure with the impure oxides. The known-a method produced biases ranging from -17.01% to -7.60%. These large biases occur because the method underpredicts a (the oxide is assumed to be pure) and thus undercorrects for multiplication. This results in an assay value that is biased high. Again for the impure oxides, the multiplicity method produces the best results with biases ranging from -2.08% to 0.84%. Note that even the low-efficiency counters produce reasonable assays, but there are differences in count times and number of samples assayed. The HLNC multiplicity assay precision is almost a factor of three worse than the 5RMC precision, even though the HLNC counted approximately twice as long.

Figure 2 is a plot of multiplicity assay bias and precision for measurements of each of the 20 pure and impure oxide items in each of the five counters. The precision of the measurements is based on the actual count times that varied from item to item and detector to detector.

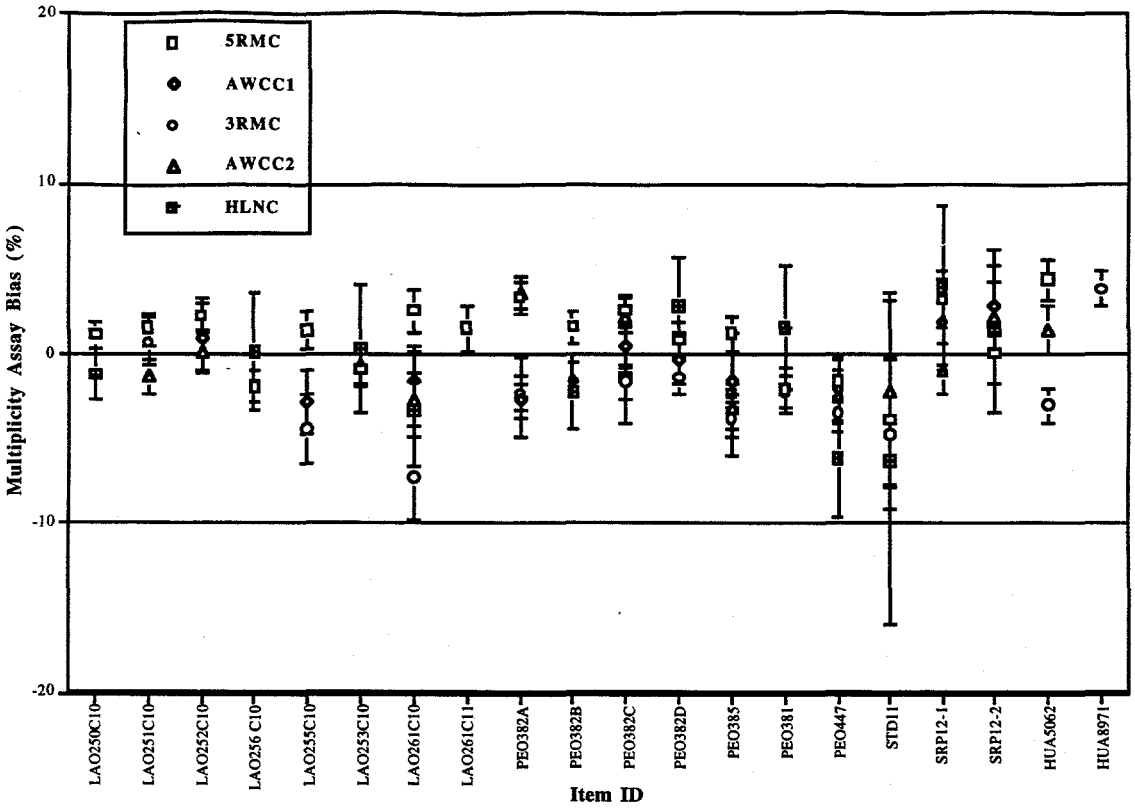

Fig. 2. Multiplicity assay bias (%) versus item ID for 5 neutron counters. Error bars reflect precision of the actual measurements.

The data from Fig. 2 show no clear bias in the measurements of individual items compared with declared values. The precisions shown for each of the measurements reflects the differences in detector efficiency and count time.

It was desired to put the five counters on a common basis for purposes of comparing measurement precisions. This was possible because four of the impure plutonium oxide items were assayed by multiplicity analysis in all five detectors. The four items were PEO382A, PEO382C, PEO385, and PEO447. These items range in plutonium mass from 20g to 875g plutonium, and their a values range from 0.7 to 0.9. The plutonium isotopic composition of these items produces ~10% effective ^{240}Pu mass fraction. The precisions of these measurements were adjusted to a count time of 1000s. Figure 3 shows a comparison plot of 1000s multiplicity assay precision versus effective ^{240}Pu mass for the four PEO impure oxide items.

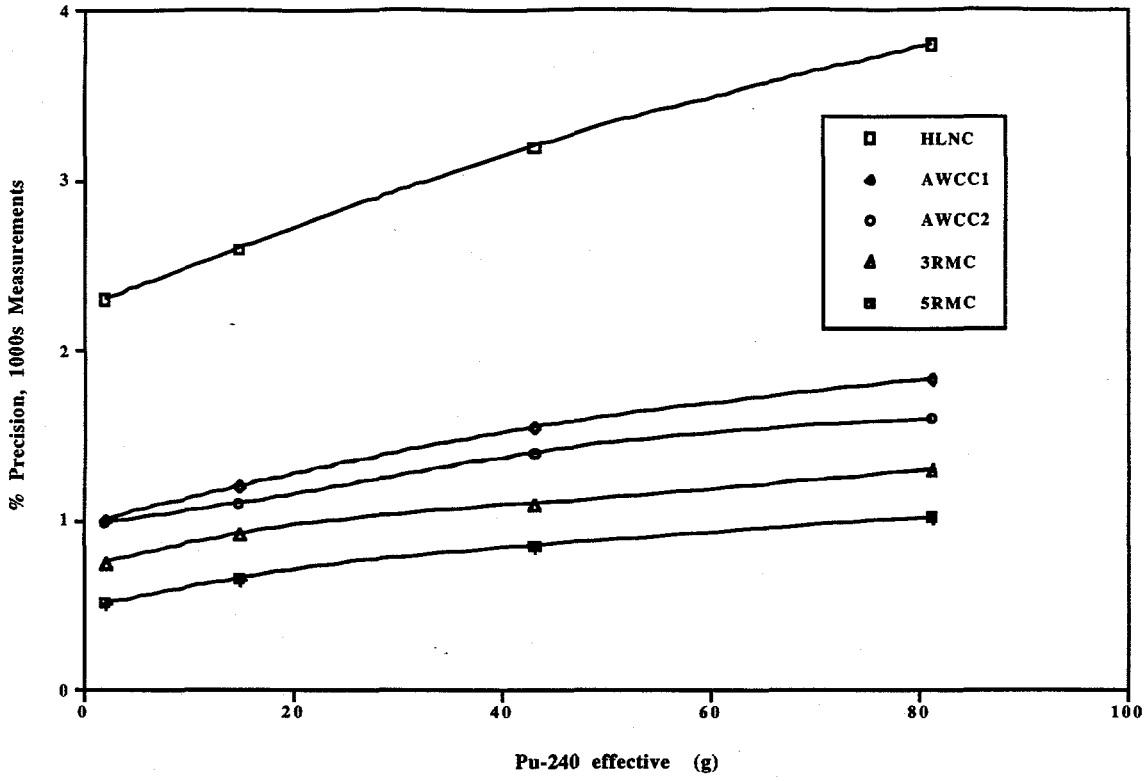

Fig. 3. 1000s multiplicity assay precision versus effective ^{240}Pu mass for four impure plutonium oxide items (PEO series, a = 0.7-0.9) and five detectors.

Fig. 3 shows that for the largest PEO item (PEO-447), the precisions are 1.02% for the 5RMC, 1.30% for the 3RMC, 1.60% for AWCC2, 1.83% for AWCC1 and 3.8% for the HLNC. These values are approximately inversely proportional to the detector efficiencies given in Table I. They also imply that to achieve the same precision, the count time required would be ~14 times as long for the HLNC as the 5RMC, but only ~2.8 times as long for the AWCCs.

The most impure of the plutonium oxide items used in this study was STD11 with an a value of ~4.3 as determined by the multiplicity assay in the 5RMC. STD11 has 59.8g plutonium and 7.66% effective ^{240}Pu mass fraction. Table IV below compares 1000s multiplicity assay precisions for STD11 in four detectors.

Table IV. Multiplicity Assay Precision for STD11 Measured in Four Detectors

Detector	% Multiplicity Assay Precision - STD11
5RMC	2.9
3RMC	4.8
AWCC2	6.4
HLNC	18.3

Table IV shows that for the multiplicity assays of STD11, which has a relatively high a value, measurement precision is highly dependent on detector type. The variation of precision is approximately inversely proportional with detector efficiency to the power 1.2-1.3. For STD11, the count time required would be ~40 times as long for the HLNC as the 5RMC, but only 4.9 times as long for the AWCC.

It was desired to compare the multiplicity assay precision of the four detector types for a nominal sample mass and a wide variation in a. A figure-of-merit[8] code was used to calculate 1000s multiplicity assay precision for a values ranging from 0.5 to 10. Results are shown in Fig. 4.

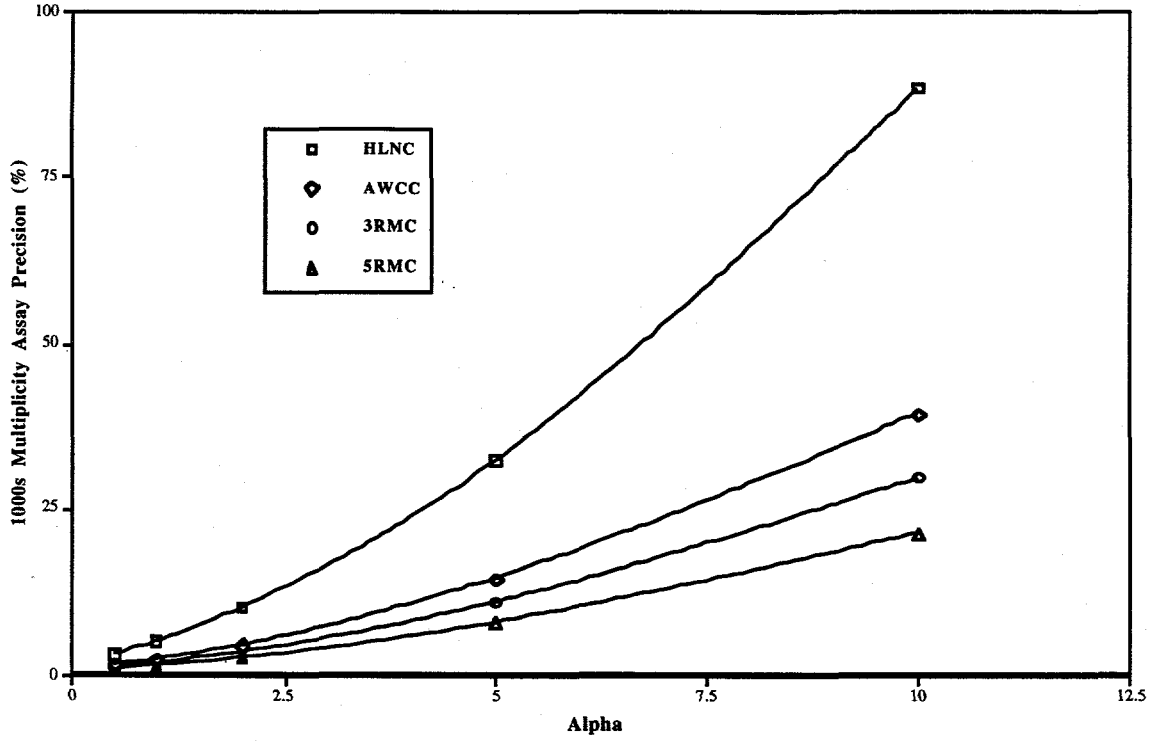

Fig. 4. 1000s multiplicity assay precision versus alpha for a sample with an effective ^{240}Pu mass of 100g (multiplication = 1.1) and for four detectors.

Fig. 4 shows the dramatic effect of increasing a in worsening the precision of multiplicity assays, especially for low efficiency detectors such as the HLNC. Even for the 5RMC, the precision for a = 10 is only 21.3%.

CONCLUSIONS

This study shows that multiplicity assays of some impure plutonium oxide and MOX items can be performed with reasonable accuracy and precision using any of the detectors employed. The primary limiting factor is counting time, and hence detector efficiency. For slightly impure items such as the PEO series (a = 0.7-0.9), even the HLNC can be used with acceptable precision in reasonable count times for partial defects type measurements. For more highly impure items such as STD11 (a = 4.3),

an AWCC could produce acceptable results in passive mode with graphite end plugs. For very highly impure items, a true multiplicity counter optimized for counting efficiency would be required. Of course, for some of these items calorimetry may be the only option.

For high-a samples, the dependence of detector efficiency on neutron energy will be more important for multiplicity assay bias than for the PEO series. This dependence is much more pronounced for the HLNC than the 5RMC.

For high-mass samples, the multiplicity deadtime of the detector must be determined accurately or multiplicity assay biases will result. Multiplicity counters are designed to have small deadtimes to reduce this problem.

These factors will hopefully aid in selection of detector options for use as multiplicity counters and also widen the applicability of the multiplicity analysis method for assay of impure plutonium oxide and MOX.

REFERENCES

1. H. O. Menlove and J. E. Swansen, "A High-Performance Neutron Time Correlation Counter," *Nuclear Technology* **71** 497-505, (November 1985).

2. H. O. Menlove, "Description and Operation Manual for the Active Well Coincidence Counter," Los Alamos Scientific Laboratory report LA-7823-M (May 1979).

3. H. O. Menlove and J. E. Stewart, "A New Method of Calibration and Normalization for Neutron Detector Families," Los Alamos National Laboratory report LA-11229-MS (April 1988).

4. M. S. Krick and J. E. Swansen, "Neutron Multiplicity and Multiplication Measurements," *Nucl. Instrum. Methods* **219**, 384-393 (1984).

5. J. E. Stewart, M. S. Krick, T. D. Reilly, D. G. Langner (LANL), W. Theis, R. Lemaire, and J. Xiao (IAEA), "Implementation of Neutron Counting Techniques at US Facilities for IAEA Verification of Excess Materials from Nuclear Weapons Production," Los Alamos National Laboratory document LA-UR-95-2329, Proc. INMM 36th Annual Meeting, Palm Desert, CA, July 1995.

6. J. E. Stewart, M. S. Krick, J. Xiao, R. J. Lemaire, V. Fotin (IAEA), L. McRae, D. Scott, and G. Westsik (WHC), "Assay of Scrap Plutonium Oxide by Thermal Neutron Multiplicity Counting for IAEA Verification of Excess Materials from Nuclear Weapons Production," Los Alamos National Laboratory document LA-UR-96-2515, Proc. INMM 37th Annual Meeting, Naples, FL, July 1996.

7. D. G. Langner, M. S. Krick, N. Ensslin, G. E. Bosler, and N. Dytlewski, "Neutron Multiplicity Counter Development," Los Alamos National Laboratory report LA-UR-91-1569, Proc.ESARDA 13th Annual Symposium, Avignon, France, May 14-16, 1991.

8. N. Ensslin, M. S. Krick, and N. Dytlewski, "Assay Variance as a Figure-of-Merit for Neutron Multiplicity Counting," Nuclear Instruments and Methods, A290 (1990) 197-207.